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Research into a number of aspects of photorefractive materials and holographic data storage is reported, including growth and characterization of optically nonlinear crystals and study of the physical mechanisms involved in holographic data storage and in permanent fixing of these holograms. We have established that ionic conductivity is the main mechanism responsible for the decay of fixed holograms in photorefractive crystals such as Lithium Niobate, and have achieved a significant increase in the hologram lifetime by reducing the OH density in the crystal by a factor of 100. The double phase conjugate mirror (DPCM) was investigated and a number of numerical studies were devoted to its detailed understanding. We demonstrated that optical beams are self-focused and self-trapped upon photopolymerization. We observed that optical interference patterns generate dynamic and remnant domain holograms in ferroelectric crystals such as SBN:75 near the ferroelectric/paraelectric phase transition.

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# **RESEARCH IN PHOTOREFRACTIVE CRYSTALS**

## **FINAL REPORT**

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## STATEMENT OF PROBLEM STUDIED:

The general area of research studied is that of holographic data storage. The main problems investigated were:

- (1) Growth and characterization of optically nonlinear crystals.
- (2) The study of physical mechanisms involved in holographic data storage and in permanent fixing of these holograms.
- (3) Theoretical and experimental studies of crosstalk in data reconstruction in holographic memories

## SUMMARY OF IMPORTANT RESULTS:

### 1. Ionic Fixing in Photorefractive Lithium Niobate.

We established that ionic conductivity is the main mechanism responsible for the decay of fixed holograms in photorefractive crystals. This decay is a primary obstacle thwarting the utilization of these crystals in archival holographic data storage. We have investigated the dependence of fixed hologram lifetime  $\tau$  in  $\text{LiNbO}_3$  (LN) on ionic conductivity and demonstrated a substantial increase in room temperature lifetimes of fixed holograms in dehydrated Fe:  $\text{LiNbO}_3$ .

The lifetime of a fixed ionic grating is determined by the conductivity  $\sigma_{\text{ion}}$  of the ionic species at a given temperature  $T$ , i.e.,  $\tau \propto \epsilon\epsilon_0/\sigma_{\text{ion}}$ . The thermally activated ionic transport obeys an Arrhenius-type dependence on the temperature  $\sigma_{\text{ion}} = e n_I \mu_0 \exp(-E_a/kT)$ , where  $n_I$  is the density of ions and  $E_a$  is the activation energy. The decay of an ionic hologram at room temperature is therefore due to residual ionic conduction, which in some cases may be slowed down by dynamic electronic screening.

We measured the decay time of holograms recorded and stored at different temperatures. We found that in as-grown LN crystals with fairly high hydrogen content (measured by the strength of the OH- stretching vibration absorption line near  $2.87\mu\text{m}$ ), the ionic conductivity exhibits 1.2 eV activation energy and is relatively strong. By extrapolation, the lifetime at room temperature ( $21^\circ\text{C}$ ) is about 70 days, which is too short for the majority of storage applications. This 1.2 eV activation energy is due to the hydrogen conductivity, which is the mechanism known to be responsible for ionic fixing in **as-grown** lithium niobate.

We achieved a significant increase in the hologram lifetime by reducing the OH<sup>-</sup> density in the crystal by a factor of 100. This was accomplished by high temperature post-growth processing in a dry oxygen atmosphere using a specially designed oxidation apparatus. We established that after substantial hydrogen removal, the ionic conductivity is determined by a species other than hydrogen. The conductivity of this carrier doesn't depend on the hydrogen content and exhibits an activation energy of ~1.4 eV. The ionic hologram lifetime in dehydrated LN crystals at room temperature is extrapolated to be ~3 years. We have also demonstrated highly efficient (~90% of initial diffraction) and long-lifetime fixing in dehydrated Fe-doped LN.

We investigated the influence of Li vacancies and deficiencies on the 1.4 eV ionic conductivity. We found that in nearly stoichiometric LN (as obtained via vapor transport equilibration in Li<sub>2</sub>O vapor) and in MgO doped LN, the extrapolated hologram lifetimes at room temperature do not exceed that obtained in congruent and dehydrated LN. This finding suggests that the most likely species associated with the 1.4 eV activation energy are lithium ions occupying the interstitial sites. We are presently conducting a detailed study in order to identify the exact origin of 1.4 eV conducting species in LN, with the goal of increasing the lifetime of fixed holograms further.

## 2. Characterization and growth of photorefractive KLTN for holographic storage

A promising photorefractive crystal for holographic data storage applications is KLTN, which is known to exhibit a strong voltage controlled photorefractive effect. We have evaluated methods to fix permanent holograms in this material. These methods require that several features of the dielectric response are accurately characterized: namely, the roles of ions, shallow electronic traps and ferroelectric domains in the space charge grating formation must be determined. Three fundamental measurements were performed:

- A. Ionic fixing is achieved at 340 K with a characteristic activation energy of 0.67 eV to 0.76 eV. The nominal diffraction efficiency of fixed ionic gratings is about 25%.
- B. Shallow trapped electrons (holes) also partially compensate the photorefractive electronic gratings. They are characterized by an activation energy of 0.12 eV. This compensating grating component can be erased by a non-Bragg matched laser beam at the same wavelength.

C. Domain switching is observed while recording holograms in the ferroelectric phase by monitoring the Barkhausen current noise. We performed this measurement under two conditions: (i) a uniform incident beam of  $1\text{W cm}^{-2}$  intensity, and (ii) two coherent, interfering beams with the same total intensity as in (i). The Barkhausen current "jumps" were observed only in case (ii), which indicates that domain switching occurred due to the optically induced space charge field alone. The lifetime of ferroelectric domain gratings agrees with the Vogel-Fulcher law. The nominal diffraction efficiency of domain gratings is as large as 55%, making this a promising material to record highly efficient volume holograms.

These measurements indicate that efficient domain and/or ionic fixing may be achieved within the same crystal, depending on the temperature of the recording process.

### 3. Double Phase Conjugate Mirror

The double phase conjugate mirror (DPCM) is a unique device in which two mutually incoherent pump beams are phase conjugated simultaneously. This device's ability to couple incoherent light sources and generate very high fidelity phase conjugate beams have made it promising for multimode to singlemode fiber-optic coupling, frequency locking of elements of a laser diode array and correlation filtering in fiber optic sensors. A number of numerical studies were devoted to its detailed understanding. We showed that the DPCM exhibits characteristics that are similar to critical phenomena and take advantage of this similarity by analyzing the DPCM through the "amplitude equation" formalism. This equation is an established approach to treat systems that exhibit critical transitions and has been successful in quantifying many of the experimental observations in these systems. Some characteristics of the DPCM that we have quantitatively analyzed are, for instance, the effects of saturation on the critical dynamics, the conjugation fidelity, the sensitivity to noise, and stability.

### 4. Self-focusing and self-trapping of optical beams upon photopolymerization

We demonstrated that optical beams are self-focused and self-trapped upon photopolymerization. This phenomenon belongs to an entirely new class of optical nonlinearity observed only at sufficiently low average optical intensity. The origin of this optical nonlinearity is the light-induced index change accompanying

photopolymerization. This permanent index change is a function of absorbed optical energy rather than intensity and is larger than the Kerr index change by orders of magnitude. While self-trapped beams in Kerr media require intensities of  $\text{MW cm}^{-2}$ , light beams can be self-trapped in photopolymers only for sufficiently **low intensities**. The response time of the Kerr nonlinearity is on the order of fs, and the nonlinear optical response time of photopolymerization is typically in excess of milliseconds. Like the Kerr effect, photopolymerization is a local phenomenon in space, for the index change at any location depends only on the light intensity at that same location. However, the index change is **nonlocal in time**, for it depends on the history of the optical electric field at all earlier times. This leads to a fundamentally new form of the nonlinear wave equation displaying transient, self-guiding solutions. In addition, a unique condition for self-trapping in photopolymers is that the average intensity must lie below an approximate threshold value given by  $I_{\text{ave}} < U_0/\tau$ , where  $U_0$  is the critical exposure to cure the photopolymer and  $\tau$  is the monomer radical lifetime. For a typical photopolymer composition, the average intensity must be less than  $10 \text{ W cm}^{-2}$ . This optical nonlinearity is unique in that it occurs only for low average optical intensity. Intuitively, if more intense illumination were used, a large number of radicals would be produced in a time which is short compared to the radical lifetime  $\tau$ . The photopolymer would then cure completely in the illuminated region, before the optical beam experienced an index change that could channel the light deeper into the material.

We used self-trapping to fabricate polymeric microstructures. We observe dramatic self-trapping of optical beams over distances in excess of 3 cm and beam diameters of 10 to 50 mm by exposing the polymer to a sequence of short exposures. Furthermore, the waveguides can be steered during their formation by introducing an asymmetry across the transverse intensity profile of the beam. An array of fiber-like structures have been fabricated to produce a three-dimensional microlattice. The diameters of the light channels remain constant well beyond the confocal parameter, a signature of self-trapping.

## 5. Optically induced domain gratings in SBN

We observed that optical interference patterns generate dynamic and remnant domain holograms in ferroelectric crystals such as SBN:75 near the ferroelectric/paraelectric phase transition. A series of optical and electrical measurements have confirmed that this new phenomenon is ubiquitous in certain

photorefractive crystals such as strontium barium niobate. This understanding allowed us to optimize and subsequently apply the technique to two technologies in nonlinear optics: tunable quasi-phase matched second harmonic generation and holographic data storage.

Three distinct observations established that ferroelectric domain gratings are formed by light. First, the waveguiding of light along the electrically erasable domain grating was revealed with a high resolution optical microscope. Second, the angular distribution of the scattered second harmonic light indicates that the nonlinear optically susceptibility and hence the local spontaneous polarization is spatially modulated. Third, temporal spikes in the displacement current along the crystalline c axis provide direct evidence that discrete domain switching is induced while recording optical holograms.

We conducted holographic experiments which isolated the roles of temperature, intensity, and time in the grating recording process. By tailoring these recording conditions, the normally transient photorefractive holograms were rendered permanent, a prerequisite for holographic data storage applications. We established that this memory arises from ferroelectric hysteresis. In a sense, holograms encoded in the remnant domain structure provide a three dimensional analog to a magnetic recording media, with an ultimate data storage density of 1 Terabyte/cm<sup>3</sup>. These holograms or pages of data can be electrically multiplexed/demultiplexed to simplify the accessing of information, which we demonstrate experimentally and analyze theoretically.

We also used domain holograms of the proper spatial periodicity to convert infrared light to visible light through a quasi-phase matched, second harmonic generation process. This technique provided a new technology for extending the wavelength range of laser sources. An in-depth theoretical and experimental overview of this work comprises Chapter 9 of "The Photorefractive Effect," Kluwer Academic (1994) and a pending US Patent (no. 80/037,076) filed in 1993.

## 6. Photorefractive crystal growth

We grew doped K<sub>1-y</sub>Li<sub>y</sub>T<sub>1-x</sub>N<sub>x</sub>O<sub>3</sub> (KLTN) crystals with 30 different compositions. The range of compositions include 0 < y < 0.15, 0.05 < x < 0.35. The dopant compositions include V,Cu; Cr,Fe; Ti,Fe; Ti,V,Ni; Ti,Ni; V,Ni; Ti,V,Co; Ti,V,Mn; Ti,V,Cu Ti,V,Zn. The resulting samples are single crystals of typically high optical quality. The goal of growing crystals with a broad range of compositions was twofold:

- i) To tailor the paraelectric/ferroelectric phase transition temperature T<sub>c</sub> (200K-350K). The optical and electrical properties display unique behavior near T<sub>c</sub> that

- may be exploited to produce long lived, high diffraction efficiency volume holograms.
- ii) To extend the photorefractive beam coupling response to lower wavelengths (i.e., from visible to near infrared ).

In addition to the growth of KLTN, we grew Sr<sub>1-x</sub>Ba<sub>x</sub>TiO<sub>3</sub> (SBT). SBT is also a promising photorefractive material, with a phase transition temperature of 250 K-450 K. The transition temperature can be tailored by changing the mol fraction of Ba (i. e., x). 10 x 10 x 3 mm single crystals of SBT have been obtained with x=0.15. This composition exhibits a ferroelectric/paraelectric phase transition near 150 °C. This material shares two desirable optical properties with KLTN: strong infrared beam coupling response and voltage controlled high diffraction efficiency. In the near future we plan to grow an SBT crystal with a larger mol fraction x. A composition with x=0.3 is expected to give a transition in the range of 0 to 50 °C.

The holographic exposure energy to saturation has been investigated by measuring the diffraction efficiency of two beam coupling. At 633 nm, with an applied electric field of 1500V cm<sup>-1</sup> at T<sub>C</sub> + 5 °C, the beam coupling is approximately 10<sup>-4</sup> cm<sup>3</sup> J<sup>-1</sup>.

## 7. Fixing of holograms in KLTN

The process by which a photorefractive holographic grating is converted to one which is not erased under illumination is referred to as fixing. The ability to fix gratings with high efficiencies and long lifetimes is essential for holographic archival data storage applications and for the fabrication of thick holographic optical elements. Thermal fixing refers to the process by which a photorefractive space charge grating is compensated at elevated temperatures by a mobile non photoactive species. At lower temperatures the sample is illuminated, partially erasing the photorefractive grating and revealing a fixed grating unaffected by illumination. Thermal fixing has been observed in ilmenite materials (LiNbO<sub>3</sub>, LiTaO<sub>3</sub>, BSKNN) and in the perovskite material KNbO<sub>3</sub>. High fixed efficiencies of 98% have been achieved in LiNbO<sub>3</sub>. However, this material exhibits low photorefractive sensitivity, making it unsuitable for many holographic data storage applications. Large values of photorefractive sensitivities have been reported in

$\text{KNbO}_3$ , however reported fixing efficiencies are less than 1%. Materials are needed with both high photorefractive sensitivity and high fixing efficiency.

We demonstrated fixing of holograms with high efficiency in a sample of KLTN. Thermal fixing occurs by screening of a photorefractive space charge field by a mobile species at elevated temperatures followed by revealing of the fixed grating under illumination at lower temperatures. Near 80% conversion of the optically written electronic grating to a fixed grating is achieved. The thermal decay of the hologram exhibited an Arrhenius type dependence with an  $E_A = 0.67 \pm 0.03$  eV activation energy. Hologram fixing has been observed in both the para and ferroelectric phases, in the temperature range of +/- 10 degrees of the Curie temperature. The fixed diffraction efficiency is very strong (typically 60%) and the lifetime of readout exceeds several hundred hours. We are presently investigating the physical origin of the long lived grating component.

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A. Kewitch, M. Segev, A. Yariv, "Permanently Fixed Volume Phase Gratings in Ferroelectrics," Chapter 9, The Photorefractive Effect, Ed. D. Nolte, Kluwer Academic, 1994.

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REPORT OF INVENTIONS:

A. S. Kewitsch and A. Yariv, "Self-trapping and self-focusing of optical beams upon photopolymerization" and "Holographically fabricated porous polymers", U.S. Provisional Patent applications; combined into non-provisional application 08/981,181.

A. S. Kewitsch and A. Yariv, U. S. Patent application 08/741,267, "Lens-like photoresists for projection photolithography."